JC09 Rec'd PCT/PTO 13 MAR 2001

FORM-PTO-1390 (Rev. 12-29-99) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER: 025219-317

U.S APPLICATION NO. (If known, see 37 C.F.R. 1.5): Unassigned 09/786985

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/FR99/02238	September 21, 1999	September 22, 1998

TITLE OF INVENTION: MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION

APPLICANT(S) FOR DO/EO/US: Patrick HOURQUEBIE; Didier MARSACQ; Philippe MAZABRAUD

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and the PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☒ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:

PCT Request, International Search Report and cited references

JC02 Rec'd PCT/PTO 13 MAR 2001

U.S. APPLICATION NO. (If known, see 37 C.F.R. 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
Unassigned 09/786985	PCT/FR99/02238	025219-317

17. ☒ The following fees are submitted:	CALCULATIONS	PTO USE ONLY
Basic National Fee (37 CFR 1.492(a)(1)-(5)):		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO $1,000.00 (960)		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO $860.00 (970)		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO $710.00 (958)		
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) $690.00 (956)		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) $100.00 (962)		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 860.00	
Surcharge of **$130.00 (154)** for furnishing the oath or declaration later than 20 ☐ 30 ☐ months from the earliest claimed priority date (37 CFR 1.492(e)).	$	

Claims	Number Filed	Number Extra	Rate		
Total Claims	18 -20 =	0	X$18.00 (966)	$ 0.00	
Independent Claims	2 -3 =	0	X$80.00 (964)	$ 0.00	
☐ Multiple dependent claim(s) (if applicable)			+$270.00 (968)	$ 0.00	
			TOTAL OF ABOVE CALCULATIONS =	$ 860.00	
Reduction for 1/2 for filing by small entity, if applicable (see below).				$	-
			SUBTOTAL =	$ 860.00	
☐ Processing fee of **$130.00 (156)** for furnishing the English translation later than 20 ☐ 30 ☐ months from the earliest claimed priority date (37 CFR 1.492(f)).			+	$	
			TOTAL NATIONAL FEE =	$ 860.00	
☐ Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **$40.00 (581)** per property +				$	
			TOTAL FEES ENCLOSED =	$ 860.00	
☐			**Amount to be: refunded**	$	
			charged	$	

a. ☐ Small entity status is hereby claimed.

b. ☒ A check in the amount of $ 860.00 to cover the above fees is enclosed.

c. ☐ Please charge my Deposit Account No. 02-4800 in the amount of $_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

d. ☐ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 02-4800. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Robert E. Krebs
BURNS, DOANE, SWECKER & MATHIS, L.L.P.
P.O. Box 1404
Alexandria, Virginia 22313-1404
(650) 622-2300

SIGNATURE

Robert E. Krebs
NAME

25,885
REGISTRATION NUMBER

Patent
Attorney's Docket No. 025219-317

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of	)	
	)	
Hourquebie, et al.	)	Group Art Unit: Unassigned
	)	
Application No.: Unassigned	)	Examiner: Unassigned
	)	
Filed: Herewith	)	
	)	
For: MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION	)	

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to examination, please amend the subject application as follows:

IN THE CLAIMS

Claim 3, line 13, please delete "or 2"

Claim 4, line 23, please delete "or 2"

Claim 5, line 2, please delete "or 2"

Claim 6, line 8, please delete "or 2"

Claim 9, line 28, please delete "such as defined in claim 1"

Claim 18, line 27, please delete "or 17"

PLEASE ADD THE FOLLOWING CLAIMS:

19. Material having improved resistance to thermal ageing according to claim 2, characterized in that the insulating polymer is chosen from among the thermoplastic resins such as acrylic, styrene, vinyl or cellulose resins, or from among polyolefins, fluorine-containing polymers, polyethers, polyimides, polycarbonates, polyurethanes, silicones, their copolymers or mixtures between homopolymers and copolymers.

20. Material having improved resistance to thermal ageing according to claim 2, characterized in that the insulating polymer is chosen from among polyethylene, low density polyethylene, high density polyethylene, linear low density polyethylene, polypropylene, ethylene-propylene-diene monomer, fluorine-containing polyvinylidene, ethylene butacrylate or the copolymers of ethylene and vinyl acetate, either alone or in a mixture.

21. Material having improved resistance to thermal ageing according to claim 2, characterized in that the insulating polymer is chosen from among the thermosetting resins, such as polyesters, epoxy resins or phenol resins.

22. Material having improved resistance to thermal ageing according to claim 2, characterized in that the conducting polymer has a conductivity of at least approximately $10^{-9}S.cm^{-1}$.

23. Use of the insulating material having improved thermal resistance obtained with the method according to any of claim 17, for the manufacture of high and/or very high voltage cables.

REMARKS

The claims of the subject application have been amended to avoid multiple dependency. Favorable consideration of the subject application is respectfully requested.

Respectfully submitted,

BURNS, DOANE, SWECKER & MATHIS, L.L.P.

By: _____
Robert E. Krebs
Registration No. 25,885

Post Office Box 1404
Alexandria, Virginia 22313-1404
(650) 622-2300

Date: March 12, 2001

H4

PCT

Patent
Attorney's Docket No. 025219-317

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of	)	
	)	
Hourquebie, et al.	)	Group Art Unit: Unassigned
	)	
Application No.: 09/786,985	)	Examiner: Unassigned
	)	
Filed: March 13, 2001	)	
	)	
For: MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION	)	

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to examination, please amend the subject application as follows:

IN THE SPECIFICATION

Please amend the specification by inserting before the first line the sentence:

"This application is a national phase of PCT/FR99/02238 which was filed on September 21, 1999 and was not published in English."

REMARKS

Entry of the foregoing amendment to the Specification is requested to comply with the requirements of 37 C.F.R. 1.78(a)(2).

If the Examiner should be of the opinion that a telephone conference would be helpful in resolving any outstanding issues, the Examiner is urged to contact the undersigned.

Respectfully submitted,

BURNS, DOANE, SWECKER & MATHIS, L.L.P.

By: _____

Robert E. Krebs
Registration No. 25,885

Post Office Box 1404
Alexandria, Virginia 22313-1404
(650) 622-2300

Date: March 29, 2001

Patent
Attorney's Docket No. 025219-317

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of	)	
	)	
Hourquebie, et al.	)	Group Art Unit: Unassigned
	)	
Application No.: 09/786,985	)	Examiner: Unassigned
	)	
Filed: March 13, 2001	)	
	)	
For: MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION	)	

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to examination, please amend the subject application as follows:

IN THE SPECIFICATION

Please amend the specification by inserting before the first line the sentence:

"This application is a national phase of PCT/FR99/02238 which was filed on September 21, 1999 and was published by the International Bureau in English on March 30, 2000."

REMARKS

Entry of the foregoing amendment to the Specification is requested to comply with the requirements of 37 C.F.R. 1.78(a)(2).

If the Examiner should be of the opinion that a telephone conference would be helpful in resolving any outstanding issues, the Examiner is urged to contact the undersigned.

Respectfully submitted,

BURNS, DOANE, SWECKER & MATHIS, L.L.P.

By: _____
Robert E. Krebs
Registration No. 25,885

Post Office Box 1404
Alexandria, Virginia 22313-1404
(650) 622-2300

Date: March 28, 2001

MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION

DESCRIPTION

Technical field

The present invention relates to a material with improved resistance to thermal ageing, to a method for producing this material, and to the use of this material in the manufacture of high and very high voltage cables.

The material according to the invention is an insulating material which has improved characteristics of resistance to thermal ageing, in particular resistance to thermal ageing by oxidation.

This material with improved characteristics of thermal ageing may be used in any device requiring electric insulation, and in particular for very high voltage cables, even at high temperatures.

Prior art

The materials used to produce electric insulation cables or devices often contain insulating organic polymers such as polyolefins.

The production of materials having improved resistance to thermal ageing, in particular to thermal ageing by oxidation, involves the addition of antioxidants.

Generally, the antioxidants used to stabilize polyolefins are molecules of fairly low molecular weight which tend to migrate towards the outside of the polymer.

The materials so produced do not resist well to ageing, in particular to thermal ageing by oxidation.

Description of the invention

The purpose of the present invention is precisely to provide a material having very high thermal stability over time, in particular against oxidation.

The material of the invention is a material with improved resistance to thermal ageing containing a conducting polymer, preferably 10 to 5000 ppm of conducting polymer, dispersed in an insulating polymer and whose heterogeneity size is 0.1 µm or less as observed under electron microscopy.

The materials of the invention contain very low levels of conducting polymer, also called conjugate polymer hereinafter, typically from 10 to 5000 ppm, in the doped or undoped state.

Advantageously, the insulating polymer may be chosen from among the thermoplastic resins such as the acrylic, styrene, vinyl or cellulose resins or from among the polyolefins, fluorine-containing polymers, polyethers, polyimides, polycarbonates, polyurethanes, silicones, their copolymers or mixtures between homopolymers and copolymers.

For example, the insulating polymer may be chosen from among polyethylene, low density polyethylene, high density polyethylene, linear low density polyethylene, polypropylene, ethylene-propylene-diene monomer, fluorine-containing polyvinylidene, ethylene butacrylate or copolymers of ethylene and vinyl acetate, either alone or in a mixture.

Document US-A-5 254 633 describes a method for producing conducting or non-conducting films or coatings of polyaniline on a substrate and items formed using said method. It sets forth that particle size is not critical and may vary widely for example from 10^{-9} to 10^{-3} cm^3.

The insulating polymer may also be a thermosetting polymer chosen from among the polyesters, epoxy resins or phenol resins.

Advantageously, the conducting polymer has a melting or softening point compatible with its use with the chosen insulating polymer. In addition, the purity of this conducting polymer must be maximum otherwise impurities may affect the voltage resistance characteristics obtained with the materials of the invention. It must be soluble in organic solvents in its doped state (oxidized) or preferably in its undoped state (reduced).

According to the invention, the conducting polymer may be a charge of organic type having a π system of electrons, relocated on at least 7 atoms, on the main chain of the polymer or the branches thereof. This conducting polymer may either be a simple conducting polymer, or a conducting polymer grafted onto an insulating polymer, or a copolymer containing one or more conjugate systems, or any organic molecule sufficiently relocated or having sufficient conductivity of at least approximately $10^{-9} S.cm^{-1}$.

Such molecules may, for example, be polypeptides or vitamin A. In respect of polymers, these are advantageously chosen in the group comprising polythiophene, polyalkylthiophenes, polyaniline, polypyrrole, polyacetylene, polyparaphenylene, their derivatives or their mixtures.

The materials of the invention are obtained by a homogeneous mixture of a conjugate polymer and a polyolefin leading to homogeneity on a scale of less than 0.1 µm. Producing such materials by direct mixing of powders or granules does not lead to obtaining an

improvement in the ageing properties with conjugate polymer levels in the order of 1000 ppm.

Therefore the invention also provides a method for producing materials having improved resistance to ageing, in particular to thermal ageing by oxidation, characterized in that it comprises the steps consisting of:

- dissolving at least one conducting polymer in an organic solvent, so as to form an impregnating solution,
- impregnating granules formed of an insulating polymer or of a mixture of insulating polymers with said impregnating solution,
- evaporating the solvent so as to obtain granules of insulating polymer coated with a conducting polymer,
- drying said granules,
- extruding or hot mixing said granules to form a homogeneous mixture.

With the method of the invention it is possible to disperse the conjugate polymer in the insulating polymer on almost molecular scale.

Producing the mixtures includes a first impregnation phase of the insulating polymer granules with a solution containing the conjugate polymer. The use of this method leads to obtaining a very good mixture having the required homogeneity, but in no way amounts to a limitation of the invention. Any other method with which it is possible to obtain homogeneous mixtures on a scale of less than 0.1 micron will be suitable.

After evaporating the solvent, the granules of insulating polymer are coated with the organic charge. The granules are then dried in a drying oven and

extruded. The strip obtained is granulated. The granules obtained may be formed by any conventional technique used for insulating polymers such as casting, rolling, injection, extrusion.

The structural characteristics of the conjugate polymer such as its conjugation length, average molecular weight, percentage defect in the production line, even at very low levels, have an effect on the thermal stabilisation obtained with the materials of the invention. These characteristics may be controlled by the conditions of synthesis of the conjugate polymer.

According to the method of the invention, the conducting polymer may represent from 10 to 5000 ppm of the insulating polymer.

According to the invention, the insulating polymer and/or the conducting polymer may be the one or those previously cited.

The material of the invention shows very good resistance to thermal ageing, in particular to oxidation, as shown by the illustrative examples below which are non-restrictive, and in the graphs of appended figures 1 and 2.

This material, obtained for example by the above-cited method, may be used in the manufacture of high and/or very high voltage cables.

More precisely, it is possible to replace chemically cross-linkable polyethylene (CCP), currently used in very high voltage cables, by the material obtained with the method of the invention.

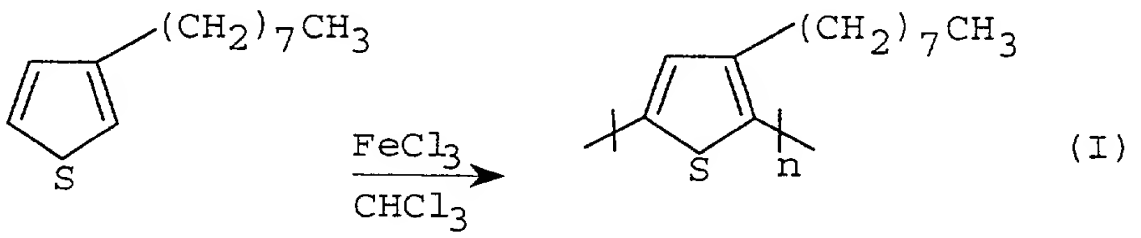
Description of the figures

Figure 1 is a graph showing the change in absorbency at 1715 cm^{-1} (acids and ketones) in relation to thermal oxidation time for a virgin polyethylene and for the same polyethylene stabilized as described in example 1 (PBT $FeCl_3$) and in example 2 (PBT Magnesium) below.

Figure 2 is a graph showing the change in absorbency at 1715 cm^{-1} (acids and ketones) in relation to thermal oxidation time for a virgin polyethylene and for a stabilised polyethylene as described in example 3 below.

Example 1

Poly(octyl-3 thiophene) (POT) was synthesized by oxidation with ferric chloride in chloroform following the method described by R. SUGIMOTO, S. TAKEDA, H.B. GU, K. YOSHINO, in Chemistry Express, vol. 1, n° 11, pp. 635-638 (1986). The following chemical equation (I) summarizes this synthesis:



(I)

The parameters affecting thermal stabilization properties here are the oxidiser over monomer ratio, the solvent and polymerization temperature.

400 mg of the above-cited polymer are dissolved in 200 ml of tetrahydrofurane (THF) so as to obtain the impregnating solution. 200 g of low density polyethylene granules are then added. The solvent is

evaporated at 50°C using a rotary evaporator. A film of conjugate polymer is then deposited on the granules. These granules are dried in a vacuum, at ambient temperature for 24 h. The granules are then extruded. A strip of mixture is obtained using a flat die with a width of 50 mm.

The material is in the form of a clear, brick-red strip containing 2000 ppm POT.

Analysis under scanning electron microscope does not detect any heterogeneity on the scale of 0.1 µm in the material produced in this example.

By way of comparison, the same material made by directly mixing the powder of conducting polymer and the powder of insulating polymer before extrusion, leads to a material whose heterogeneity size is in the order of 0.2 micron.

Measurements of oxidation rate during ageing at 80°C of the material produced in this example were taken at the same time as measurements of oxidation rate during ageing of a virgin polyethylene at 80°C. Ageing was conducted in an oven.

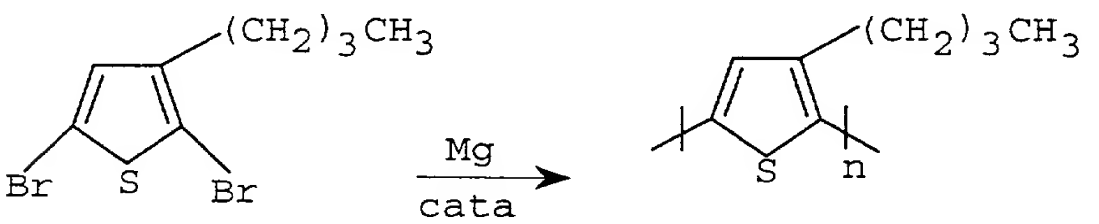
Figure 1 is a graph plotted using these measurements showing the change in absorbency at 1715 cm^{-1} (acids and ketones) in relation to the time of thermal oxidation in hours for a virgin polyethylene (graph referenced 1) and the same polyethylene stabilized as described in this example 1 (graph referenced 3).

It will be noted from these results, in comparison with the virgin polyethylene, that there is a reduction in oxidation rate during ageing at 80°C for the material of the invention. It is therefore shown that

with the presence of the conjugate polymer it is possible to stabilize the material.

Example 2

Poly(butyl-3 thiophene) (PBT) was synthesized using the general method described by O.INGANAS, W.R. SALANEK, J.E. ÖSTERHOLM, J. LAAKSO in Synthetic Metals, 22, pp. 395-406 (1988) replacing iodine by bromine. The following chemical equation (II) summarizes this synthesis:



(II)

Control over synthesis conditions can achieve variation in the characteristics of the material and the final properties of the mixture. For example, molecular weight is dependent upon synthesis conditions and modifies the solubility properties of the conjugate polymer in the matrix in which it is inserted.

The polymer obtained is extruded as in example 1. A clear, orange-coloured strip is obtained containing 2000 ppm of PBT.

Analysis under scanning electronic microscope does not reveal any heterogeneity on the scale of 0.1 µm in the material produced in this example.

Measurements were made of the oxidation rate during ageing at 80°C of the material produced in this example. The results of these measurements were used to plot graph 5 in figure 1 previously cited.

From these results it is noted that, in comparison with the virgin polyethylene, there is a reduction in

oxidation rate in relation to time during the ageing at 80°C of the material of the invention. It is therefore demonstrated that with the presence of the conjugate polymer it is possible to stabilize the material. This stabilisation is of the same order as that obtained with the polymer of example one (graph denoted 3 in figure 1).

Examples 1 and 2 therefore show that conjugate polymers derived from thiophene according to the present invention can considerably delay oxidation of the polyethylene. The non-stabilized material starts to deteriorate after 1000 oven hours at 80°C, whereas after 6000 hours the materials of the invention are still not deteriorated at this same temperature.

Example 3

A mixture containing polyaniline (Pani) and low density polyethylene is made. Polyaniline powder doped with dodecylbenzenesulfonic acid is used, whose production is described for example by Y. CAO, P. SMITH, A.J. HEEGER in Synthetic Metals, 48, pp. 91-97 (1992). This powder is placed in solution in xylene. This solution is used to impregnate the granules following the method of the invention, as in example 1.

The granules obtained are extruded as in example 1. A clear, green strip is obtained containing 500 ppm of Pani.

Analysis under a scanning electron microscope does not detect any heterogeneity on the scale of 0.1 µm in the material produced in this example.

Measurements were made of the change in absorbency at 1715 cm^{-1} (acids and ketones) in relation to the time of thermal oxidation in hours (T=90°C, thickness

500 µm) for a virgin polyethylene and a polyethylene stabilised as in this example 3. Appended figure 2 is a graph plotted using these measurements.

The stabilisation of the properties obtained is also of great importance, for harsher ageing, since oxidation of the material of the present invention (graph denoted 7) only occurs on and after 600 hours' ageing at 90°C, whereas oxidation of the virgin polyethylene starts at 50 hours (graph referenced 9).

Example 4: Manufacture of a high voltage cable

The cable manufactured comprises a conductor core successively coated with an inner semiconductor shield, a material of the invention such as in examples 1 to 3, an outer semiconductor shield and a protective sheath. This cable offers better resistance to ageing by thermal oxidation, even at temperatures of 90°C.

The use of a conjugate polymer in homogeneous mixtures on a scale of 0.1 µm or less, which is the subject of the present invention, provides a solution which, in unexpected manner, leads to increasing performance as regards the lifetime of the material subjected to thermal stress.

English translation of the amended sheets of International Preliminary Examination Report

CLAIMS

1. Method for producing insulating materials having improved resistance to thermal ageing, characterized in that it comprises the steps consisting of:

- dissolving at least one conducting polymer having maximum purity in an organic solvent, so as to form an impregnating solution,
- impregnating granules, formed of an insulating polymer or of a mixture of insulating polymers, with said impregnating solution,
- evaporating the solvent so as to obtain granules of insulating polymer coated with a conducting polymer,
- drying said granules,
- extruding or hot mixing said granules to form a homogeneous mixture.

in which the conducting polymer represents 10 to 5000 ppm of insulating polymer.

2. Production method according to claim 1, characterized in that the impregnation of the granules is made by dipping the latter in the impregnating solution.

3. Production method according to claim 1, characterized in that the insulating polymer is chosen from among the thermoplastic resins such as the acrylic, styrene, vinyl or cellulose resins, or from among polyolefins, fluorine-containing polymers, polyethers, polyimides, polycarbonates, polyurethanes, silicons, their copolymers or mixtures between homopolymers and copolymers.

4. Production method according to claim 1, characterized in that the insulating polymer is chosen from among polyethylene, low density polyethylene, high density polyethylene, linear low density polyethylene, polypropylene, ethylene-propylenediene monomer, fluorine-containing polyvinylidene, ethylene butacrylate or the copolymers of ethylene and vinyl acetate, either alone or in a mixture.

5. Production method according to claim 1, characterized in that the insulating polymer is chosen from among the thermosetting resins such as polyesters, epoxy resins or phenol resins.

6. Production method according to claim 1, characterized in that the conducting polymer has a conductivity of at least approximately $10^{-9}S.cm^{-1}$.

7. Production method according to claim 6, characterized in that the conducting polymer is a simple conducting polymer, a conducting polymer grafted onto an insulating polymer, or a copolymer containing at least one conjugate system.

8. Production method according to claim 6, characterized in that the conducting polymer is chosen from among polythiophene, the polyalkylthiophenes, polyaniline, polypyrrole, polyacetylene, polyparaphenylene, their derivatives or their mixtures.

9. Material obtained with the method according to any of claims 1 to 8.

10. Use of the insulating material having improved thermal resistance obtained with the method according to any of claims 1 to 8, for the manufacture of high and/or very high voltage cables.

ABSTRACT OF THE DISCLOSURE

MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION

The present invention relates to a material having improved resistance to thermal ageing, to a method for producing this material, and to the use of this material for the manufacture of high and very high voltage cables.

The material having improved resistance to thermal ageing contains a conducting polymer dispersed in an insulating polymer and whose heterogeneity size is 0.1 μm or less as observed under scanning electron microscopy.

The method of the invention with which it is possible to obtain this homogeneity comprises the steps consisting of:

- dissolving a conducting polymer in an organic solvent, to form an impregnating solution,

- impregnating granules formed of an insulating polymer, or of a mixture of insulating polymers, with said impregnating solution.

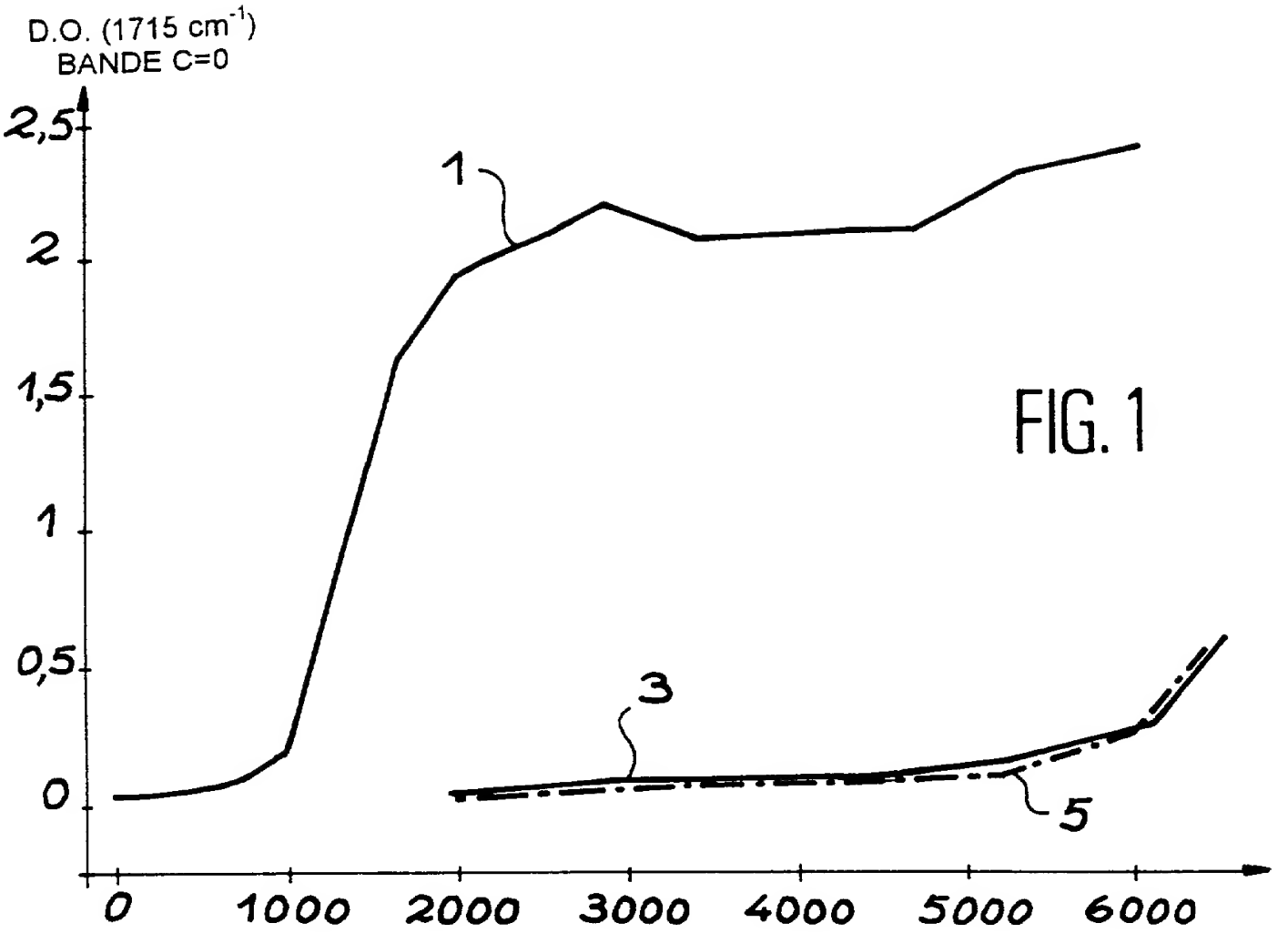

D.O. (1715 cm-1)
BANDE C=0
2,5
2
1,5
1
0,5
0
1
3
5
0
1000
2000
3000
4000
5000
6000
DUREE EN HEURES

FIG. 1

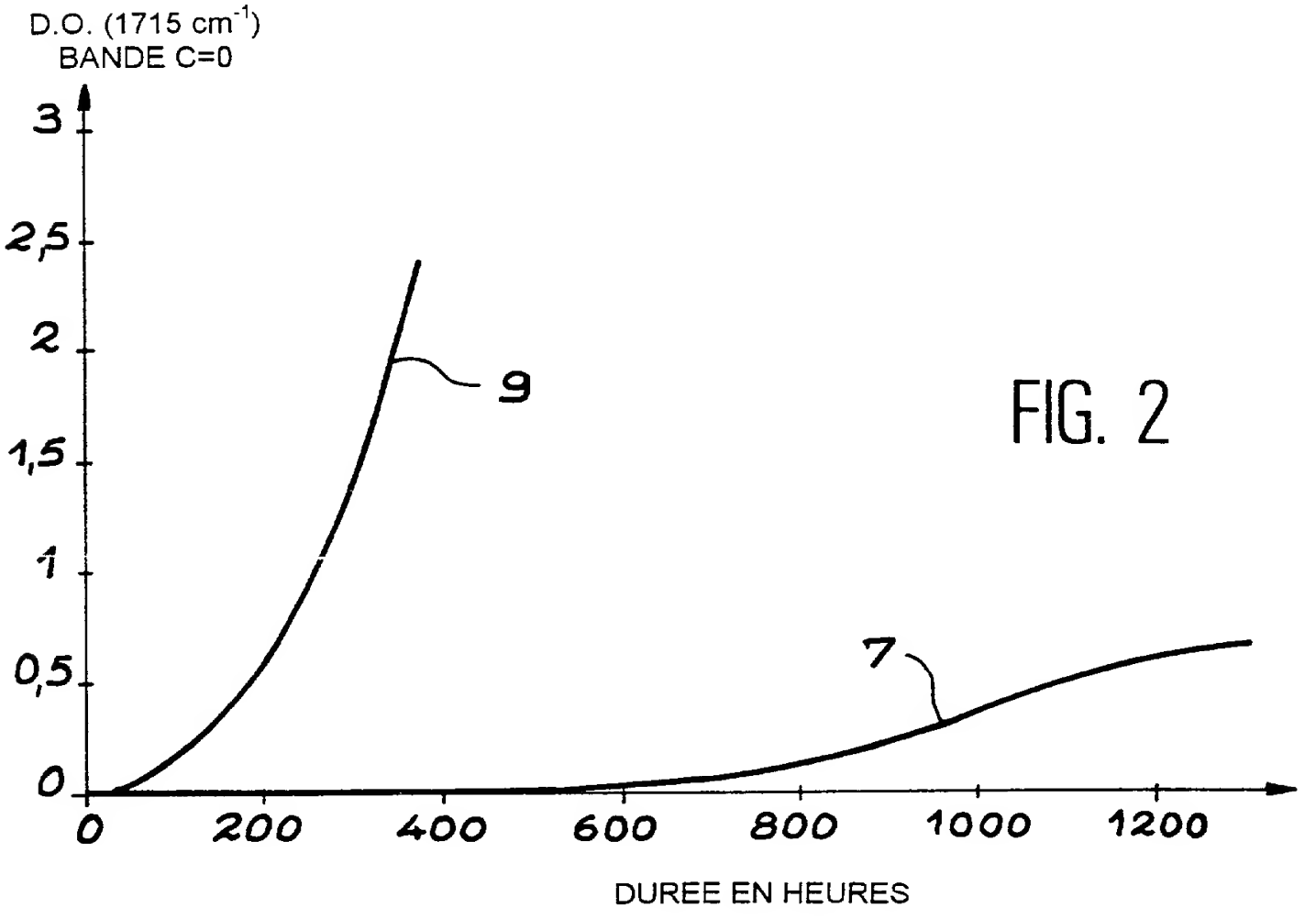

D.O. (1715 cm-1)
BANDE C=0
3
2,5
2
1,5
1
0,5
0
9
7
0
200
400
600
800
1000
1200
DUREE EN HEURES

FIG. 2

B 13019.3 EE

Declaration, Power Of Attorney and Petition

WE (I) the undersigned inventor(s), hereby declare(s) that :

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

MATERIAL WITH IMPROVED RESISTANCE TO THERMAL AGEING AND ITS METHOD OF PRODUCTION

the specification of which

☐ is attached hereto.

☒ was filed on March 13, 2001

as Application Serial No. 09/786,985

and amended on

☒ was filed as PCT international application

Number PCT/FR99/02238

on September 21, 1999

and was amended under PCT Article 19

on september 25, 2000 and November 29, 2000

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119 (a)-(d) or § 365 (b) of any foreign application(s) for patent or inventor's certificate, or § 365 (a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application (s)

Application No.	Country	Day/month/Year	Priority Claimed
98 11786	FRANCE	22 SEPTEMBER 1998	☒ YES ☐ NO
			☐ YES ☐ NO
			☐ YES ☐ NO
			☐ YES ☐ NO

We (I) hereby claim the benefit under Title 35, United States Code, § 119 (e) of any United States provisional application(s) listed below.

(Application Number)	(Filing Date)
(Application Number)	(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. §120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of prior application and the national or PCT International filing date of this application.

Application Serial No.	Filing Date	Status (pending, patented, abandoned)

And we (I) hereby appoint : William L. Mathis, Registration Number 17,337; Robert S. Swecker, Registration Number 19,885, Platon N. Mandros, Registration Number 22,124; Benton S. Duffett Jr., Registration Number 22,030; Norman H. Stepno, Registration Number 22,716; Ronald L. Grudziecki, Registration Number 24,970; Frederick G. Michaud Jr, Registration Number 26,003; Alan E. Kopecki, Registration Number 25,813; Regis E. Slutter, Registration Number 26,999; Samuel C. Miller III, Registration Number 27,360; Robert G. Mukai, Registration Number 28,531; George A. Hovanec, Jr. Registration Number 28,223; James A. Labarre, Registration Number 28,632; E. Joseph Gess, Registration Number 28,510; R. Danny Huntington, Registration Number 27,903; Eric H. Weisblatt, Registration Number 30,505; James W. Peterson, Registration Number 26,057; Teresa Stanek REA, Registration Number 30,427; Robert E. Krebs, Registration Number 25,885; William C. Rowland, Registration Number 30,888; T. Gene Dillahunty, Registration Number 25,423; Patrick C. Keane, Registration Number 32,858; Bruce J. Boggs, Jr. Registration Number 32,344; William H. Benz, Registration Number 25,952; Peter K. Skiff, Registration Number 31,917; Richard J. McGrath, Registration Number 29,195; Matthew L. Schneider, Registration Number 32,814; Michael G. Savage, Registration Number 32,596; Gerald F. Swiss, Registration Number 30,113; Michael J. Ure, Registration Number 33,089; Charles F. Wieland III, Registration Number 33,096; Bruce T. Wieder, Registration Number 33,815; Todd R. Walters, Registration Number 34,040; Ronni S. Jillions, Registration Number 31,979; Harold R. Brown III, Registration Number 36,341; Allen R. Baum, Registration Number 36,086; Steven M. Du Bois, Registration Number 35,023; Brian P. O'Shaughnessy, Registration Number 32,747; Kenneth B. Leffler, Registration Number 36,075 and Fred W. Hathaway, Registration Number 32,236 our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of BURNS, DOANE, SWECKER & MATHIS LLP, whose post Office Address is : 1737 King Street #400, Alexandria, Virginia 22314-2727.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true ; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of the application or any patent issuing thereon.

HOURQUEBIE Patrick

NAME OF FIRST SOLE INVENTOR

Signature of Inventor

02.05-2001

Date

Residence : 9, les Magnolias
37 320 Esvres sur Indre
FRANCE

Citizen of : FRANCE

Post Office Address : The same as residence

MARSAÇQ Didier

NAME OF SECOND INVENTOR

Signature of Inventor

le 15 avril 2001

Date

Residence : 9 RUE ADRIEN RICARD

38000 GRENOBLE

FRANCE FRE

Citizen of : FRANCE

Post Office Address : The same as residence

MAZABRAUD Philippe

NAME OF THIRD INVENTOR

Signature of Inventor

le 02 mai 2001

Date

Residence : 27 bld Alexandre MARTIN

45000 ORLEANS

FRANCE FRE

Citizen of : FRANCE

Post Office Address : The same as residence

NAME OF FOURTH INVENTOR

Signature of Inventor

Date

Residence :

Citizen of :

Post Office Address : The same as residence

NAME OF FIFTH INVENTOR

Signature of Inventor

Date

Residence :

Citizen of :

Post Office Address : The same as residence